

1989 SPRING
TECHNICAL MEETING



CENTRAL STATES SECTION
The Combustion Institute
COMBUSTION FUNDAMENTALS
AND APPLICATIONS

April 30 - May 2, 1989

Ritz Carlton Hotel
Dearborn, Michigan

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TUESDAY, MAY 2, 1989

Session B-4 Diagnostics
Chairperson: Lajos Rimai, Ford Research Staff

Time	Paper No.	Title & Author	Page No.
1:00	44	Simultaneous Measurement of Mixture Fraction and Hydroxyl Radical Concentration in a Lifted Hydrogen Flame - R. S. Barlow and R. W. Dibble, Sandia National Laboratories, D. C. Fourquette, Wellesley College.....	207
1:20	45	Image Persistence and Nonlinearity of Intensified Photodiode Array Detectors and Their Effect on CARS Derived Temperatures - D. R. Snelling, G. J. Smallwood, and R. A. Sawchuk, National Research Council.....	213
1:40	46	Laser-Induced Fluorescence Measurements of OH in Laminar $C_2H_6-O_2-N_2$ Flames at High Pressure - Campbell D. Carter, Galen B. King and Normand M. Laurendeau, Purdue University.....	219
2:00		Break	
2:20	47	Measurement of Atomic Sodium in a Premixed Atmospheric Flame by Asynchronous Optical Sampling - Gregory J. Fiechtner, Galen B. King, and Normand M. Laurendeau, Mechanical Engineering and Ronald J. Kneisler and Fred E. Lytle, Chemical Engineering, Purdue University.....	225
2:40	48	Interrogation of a High Pressure Injector - Glenn Bower and David Foster, University of Wisconsin-Madison.....	231
3:00	49	Image Analysis of High Pressure Sprays - M. S. El-Beshbeeshy and M. L. Corradini, University of Wisconsin-Madison.....	237
3:20	50	Vibrational Raman Scattering Measurement of a Hydrogen-Air Flame Using a Broadband KrF Excimer Laser - J. A. Wehrmeyer, T. S. Cheng, and R. W. Pitz, Vanderbilt University.....	243

TUESDAY, MAY 2, 1989

Session B-3 Diagnostics Chairperson: Paul E. Sojka, Purdue University
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Time	Paper No.	Title and Author
9:20	38	Errors Associated with Laser Doppler Velocimeter and Phase/Doppler Diameter Measurements in Combusting Flows - J. B. Ghandhi and J. K. Martin, University of Wisconsin-Madison..... 171
9:40	39	Simultaneous Measurement of Droplet Size and Velocity in a Transient Diesel Fuel Spray - J. Y. Koo and J. K. Martin, University of Wisconsin-Madison..... 177
10:00		Break
10:20	40	On the Use of Dynamic Light Scattering to Study Multimodal Size Distributions in Flame Systems - D. T. Venizelos and T. T. Charalampopoulos, Louisiana State University..... 183
10:40	41	Photoacoustic Absorption Spectroscopy for Detection of Unburned Carbon in Exhaust Gas of Coal-Fired Combustors - Alan Dona and Robert C. Brown, Iowa State University..... 189
11:00	42	An Analysis of Soot Particle Property Effects on Laser Diagnostics and Radiation Calculation - Jerry C. Ku, Wayne State University..... 195
11:20	43	Wavelength Dependence of the Optical Properties of Soot From In-Situ Measurements - H. Chang and T.T. Charalampopoulos, Louisiana State University.. 201
11:40		Lunch

MEASUREMENT OF ATOMIC SODIUM IN A PREMIXED ATMOSPHERIC FLAME BY ASYNCHRONOUS OPTICAL SAMPLING

Gregory J. Fiechtner, Galen B. King and Normand M. Laurendeau
Flame Diagnostics Laboratory
School of Mechanical Engineering

and

Ronald J. Kneisler and Fred E. Lytle
Department of Chemistry
Purdue University
West Lafayette, IN 47907

1. Introduction

Absolute number density measurements in flames are difficult to derive from laser-induced fluorescence (LIF) signals because the rate of fluorescence quenching due to collisional deexcitation is not well known.¹ The quenching rate can be calculated through knowledge of the rate constants for individual collision partners and their respective number densities.^{2,3} But the data necessary for these calculations is available only for a limited number of molecular species, and typically has been obtained under different conditions of temperature, pressure and stoichiometry. Laser-saturated fluorescence (LSF) can be used to avoid the quenching dependence⁴ but this technique is limited by the number of molecules that can be effectively saturated. Calibration techniques must also be used to take into account incomplete saturation in the wings of the laser beam.^{5,6}

In flames above 1 atm, measurement of number density requires that picosecond lasers be used to directly determine the rapid quenching rates. Previous investigators have used picosecond lasers to do time-resolved LIF experiments on OH at atmospheric pressure using either a streak camera^{7,8} or a time-correlated single-photon counting scheme.⁹ Although these experiments have the temporal resolution necessary to obtain the needed quenching rates, the amount of time necessary to obtain the data is much greater than the time scale of turbulence. This is in part due to the repetition rate of the lasers which ranges from 1 Hz to 1 kHz. In addition, the slow triggering rate of a streak camera and the long time for photon counting make these detection schemes undesirable for turbulent measurements.

In this paper we present a new combustion diagnostic suitable for measurements in turbulent, high-pressure flames. We demonstrate the efficacy of the method via measurements of atomic sodium in a laminar premixed flame at atmospheric pressure. The technique, Asynchronous Optical Sampling (ASOPS), is a pump/probe method; however, it overcomes many of the temporal problems inherent to that method.^{10,11} Furthermore, corrections for the effects of quenching can be obtained on a time-scale necessary for practical combustion measurements.^{12,13} In the ASOPS method, rather than using a single Nd:YAG laser to construct both the pump and probe beams and an optical delay line to control the relative timing between the two pulses, two Nd:YAG lasers are used to separately generate the pump and probe beams. Central to the method is the fact that the two Nd:YAG lasers are mode-locked at slightly different frequencies. The mode-locking frequencies of these two lasers are carefully controlled to maintain a constant beat frequency, creating a periodic relative phase walk-out between the pump and the probe lasers. This has the same effect as varying the optical delay line in the conventional pump/probe method; however, the period of time needed to observe the population decay of an excited energy level can be reduced from several minutes to less than a msec.

2. Equipment and Procedure

The process is illustrated in Fig. 1a, which shows the excited state population produced by several pump pulses and the temporal position of several probe pulses. Each successive probe pulse is delayed in time (relative to the pump pulse train) by a constantly increasing duration which is determined by the beat frequency of the system.

Thus, each probe pulse samples the excited state population at a slightly later time than the preceding probe pulse. This is equivalent to varying the optical delay in a conventional instrument. The sampling process repeats itself when the cumulative delay equals the period of the pump laser. Hence, any modulation of the probe beam, resulting from the creation and subsequent decay of the excited state, repeats at the beat frequency of the system. Therefore, in contrast to a conventional pump/probe instrument, there is no need to modulate the amplitude of either beam to employ synchronous detection.

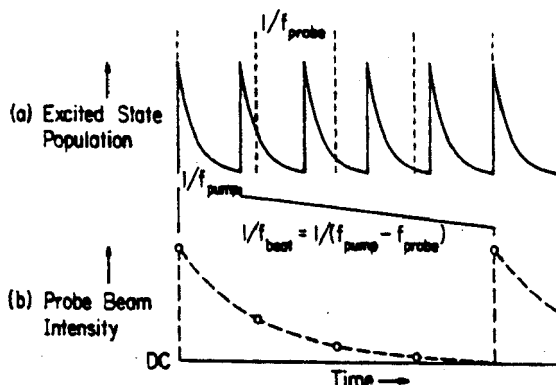


Fig. 1. ASOPS timing diagram showing (a) excited state population and (b) probe beam intensity. The probe pulses in (a) are indicated by the vertical dashed lines.

Fig. 1b illustrates the change in probe intensity which results from stimulated emission from the excited state population shown in Fig. 1a. The net effect of the ASOPS technique is that a small amplitude waveform, which is directly related to the fluorescence decay of the species under study, is impressed onto the probe laser intensity. In essence, a temporal transformation of the excited state decay is performed with the time scaled by the factor $(f_{\text{pump}}/(f_{\text{pump}} - f_{\text{probe}}))$ where f is the repetition rate of the two lasers. The ASOPS technique is thus an optical analogue of the sampling oscilloscope.

The beat frequency of the ASOPS instrument is determined by the difference in laser repetition rates or $f_{\text{beat}} = f_{\text{pump}} - f_{\text{probe}}$. The period of the pump laser, $1/f_{\text{pump}}$, determines the free temporal range, which is the maximum time available for a single decay of the excited state population. The inverse of the beat frequency, $1/f_{\text{beat}}$, represents the collection time for a single decay. The total number of points sampled during the decay is determined by $f_{\text{probe}}/f_{\text{beat}}$. The temporal difference between each sampled point within the decay profile is the sampling interval. The sampling interval and thus the temporal resolution is determined by the difference in laser periods, i.e., $f_{\text{beat}}/(f_{\text{probe}} f_{\text{pump}})$. Since the ASOPS signal follows a simple exponential, the duty cycle of the measurement is determined by the excited state lifetime of the species being monitored and the free temporal range. If we assume that the signal is zero after 3τ , where τ is the excited state lifetime, then the percent duty cycle is given by $100(3\tau f_{\text{pump}})$.

Table 1 gives the above operating parameters for various beat frequencies and laser repetition rates. Several factors must be taken into account to determine the optimum ASOPS parameters. First, to make measurements on the time-scale of turbulence, it is necessary to collect enough data for adequate signal averaging in ~ 1 msec. This criterion indicates that we should use the largest possible beat frequency. However, the beat frequency is limited by the physical cavity length of the lasers and the natural acoustic resonance frequencies of the mode-locking prisms. In addition, since the ASOPS signal is carried on the probe beam, the beat frequency must be chosen so as not to coincide with any noise in the frequency spectrum of the laser.

A second factor important to system optimization concerns the sampling interval, which must be small enough so that the ASOPS measurement has sufficient temporal resolution to reconstruct the excited state decay. This criterion sets a natural upper limit to the beat frequency because the sampling interval is proportional to f_{beat} . Finally, the duty cycle should be as close to 100% as possible. As previously shown, the duty cycle increases with the pump laser repetition rate and is independent of the beat frequency. The pump laser repetition rate can be increased by a technique called third-harmonic mode-locking. As

Table 4.1. Operating Parameters for Various Beat Frequencies.

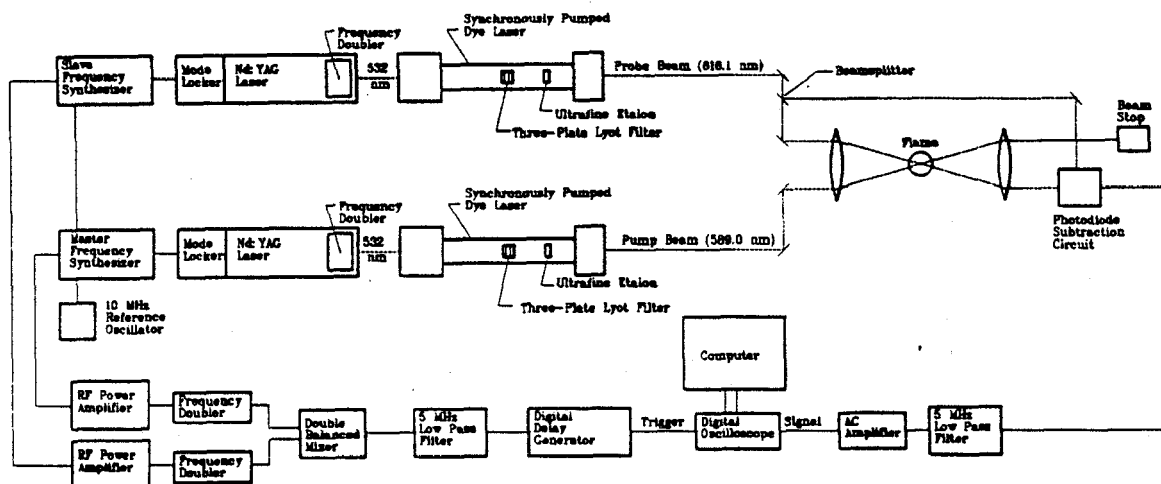
Repetition Rate (f_{pump})	~ 82 MHz	~ 82 MHz	~ 246 MHz
Beat Frequency (f_{beat})	10 kHz	100 kHz	1 MHz
Free Temporal Range ($1/f_{\text{pump}}$)	12.2 nsec	12.2 nsec	4.1 nsec
Collection Time ($1/f_{\text{beat}}$)	100 μ sec	10 μ sec	1 μ sec
Samples per Decay ($f_{\text{probe}}/f_{\text{beat}}$)	8,200	820	246
Sampling Interval ($f_{\text{beat}}/f_{\text{probe}} f_{\text{pump}}$)	1.5 psec	15 psec	17 psec
Percent Duty Cycle (for $\tau = 1$ nsec)	25%	25%	74%

can be seen from Table 1, this approach would provide the ideal case for ASOPS. The increased repetition rate not only improves the duty cycle, but also allows for higher beat frequencies and thus SNR because of the salutary effect of f_{pump} on the sampling interval.

Since the ASOPS technique requires that the pump and probe lasers operate at slightly different repetition rates, the instrument must be constructed from two independent mode-locked laser systems. Both the pump and probe beams are currently derived from Spectra-Physics model 375B dye lasers, which are synchronously-pumped by frequency-doubled, mode-locked Spectra-Physics series 3000 Nd:YAG lasers. The mode-locking frequencies are generated by two Programmed Test Sources (PTS) model 160 frequency synthesizers (accurate to 0.1 Hz) operated in a master-slave (i.e., phase-locked) configuration to minimize drift in the beat frequency of the system. Both the pump and probe beams consist of an ~ 82 MHz train of pulses, tunable (using Rhodamine 6G) from 560 to 640 nm. Three plate Lyot (birefringent) filters and ultrafine etalons placed in the dye laser cavities result in ~ 20 psec pulses with nearly transform limited bandwidths of 30 GHz¹⁴ ($\Delta\lambda = 0.04$ nm) for both the pump and probe lasers. Throughout the center of the tuning range (560 - 640 nm), average powers in excess of 120 mW are obtained using 800-900 mW of pumping power.

A block diagram of the basic ASOPS instrument is shown in Fig. 2. To obtain the trigger signal, a synchronous voltage output from each synthesizer is first amplified with an RF power amplifier (Electronic Navigation Industries 503L) and then electronically mixed by a double-balanced mixer (Anzac MDC-161). Note that the output frequency of the synthesizer is half that of the optical repetition rate; for this reason, electronic frequency doublers are included before the double-balanced mixer to obtain a trigger signal at the beat frequency of the system. The mixer output passes through a 4LM5-3-CD Texscan 5-MHz, low-pass filter to remove any high frequencies leaking through the mixer. The output of the filter then enters a digital delay generator (Berkeley Nucleonics Corporation 7095) which produces a very stable 0 to +5 V trigger pulse of variable width. Unlike the optical triggering scheme previously employed,^{11,15} this new electronic scheme offers much less temporal jitter in the trigger pulse and thus an improved SNR.

The pump and the probe beams are first passed through calibrated neutral density filters which provide a convenient means of varying the power in either beam. The two beams then pass through a single focusing lens ($f = 100$ mm) and cross at an included angle of $\sim 5^\circ$ in the flame. The probe beam is detected using the circuit of Fig. 3. Here, approximately half of the probe is split off before the flame and reaches one of the photodiodes (EG&G FND-100Q). After the flame, the probe is recollimated and monitored by the second photodiode. Reduction of probe-beam noise of up to 20 dB results from the high common-mode rejection ratio of the AD 521 instrumentation amplifier. The output of the detection circuit is filtered with a 4LM5-3-CD Texscan 5-MHz low-pass filter to remove the high frequency noise. The output from the filter is amplified by a C-COR 4375-A wideband AC amplifier by a factor of 100. The output from the



amplifier is then directed to a digitizing oscilloscope (Hewlett-Packard 54100A), triggered at the beat frequency of the system. Currently, the ASOPS repetition rates are $f_{\text{pump}} = 81.59358620$ MHz and $f_{\text{probe}} = 81.58362200$ MHz, giving a beat frequency $f_{\text{beat}} = 9.964$ kHz. The interaction volume of the pump and the probe beams is ~ 90 pL in the flame.

3. Results and Discussion

As the first substantive test of ASOPS in a combustion environment, we have chosen to detect atomic sodium that has been atomized into a premixed $\text{C}_2\text{H}_4/\text{O}_2/\text{N}_2$ flame. The flame is supported by a water-cooled, 31-mm diameter Hastelloy X flat flame burner. A NaCl solution is directed with a multistatic pump to a nebulization chamber, after which mist droplets are carried to the burner via the combined O_2/N_2 flow. Fig. 4 shows the ASOPS signal for sodium in an atmospheric flame. The pump beam is set to the sodium D_2 ($3\text{S}_{1/2} \rightarrow 3\text{P}_{3/2}$) transition (589.0 nm) while the probe beam is set to the $3\text{P}_{3/2} \rightarrow 5\text{S}_{1/2}$ transition (616.1 nm). Thus, the ASOPS signal monitors the population of the $3\text{P}_{3/2}$ state, which modulates the probe beam intensity through excited-state absorption.

The decay curve in Fig. 4 was obtained by averaging over 256 separate single-decay curves. For alkali atoms such as sodium, the decay of the $3P_{3/2}$ state is not a true exponential, but is governed by a second-order decay incorporating the quenching rate ($3P_{3/2} \rightarrow 3S_{1/2}$) and the doublet mixing rates ($3P_{3/2} \approx 3P_{1/2}$).^{16,17} A curve fit to the data with a .05% standard error is shown in Fig. 5, resulting in a $3P_{3/2} \rightarrow 3S_{1/2}$ quenching time of 830 psec and a $3P_{3/2} \rightarrow 3P_{1/2}$

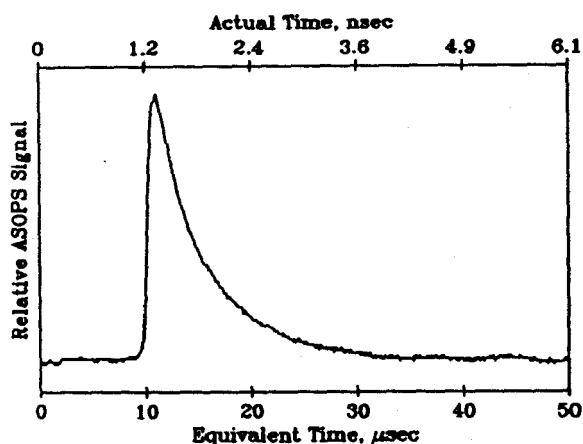


Fig. 4. Temporal decay for atomic sodium obtained with a 589.6-nm pump beam (120 mW) and a 616.1-nm probe beam (3 mW). The lower scale gives the equivalent time displayed on the oscilloscope, while the upper scale gives the actual time.

mixing time of 500 psec. This compares to lifetimes of 710 psec and 330 psec reported by Takubo *et al.*¹⁷ for sodium in a propane-air flame.

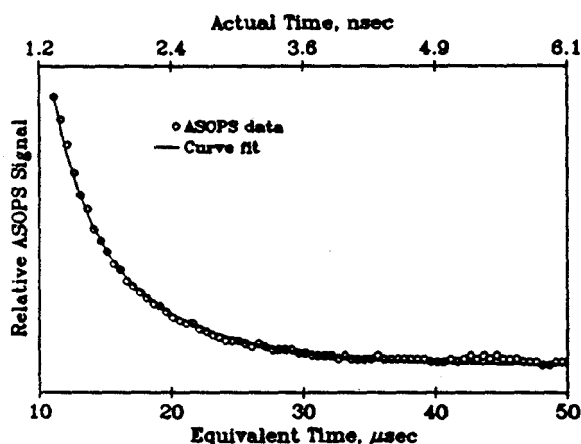


Fig. 5. Second order curve fit to the decay of Fig. 4. A $3P_{3/2} \rightarrow 3S_{1/2}$ quenching rate of $1.2 \times 10^9 \text{ sec}^{-1}$ and a $3P_{3/2} \rightarrow 3P_{1/2}$ doublet mixing rate of $2.0 \times 10^9 \text{ sec}^{-1}$ result in a fit (indicated by the solid curve) with .05% relative error from the experimental results (data points indicated by circles). To distinguish the curve fit from the experimental data, only every fifth data point is plotted.

The results of Fig. 4 represent a factor of 2.5 improvement in peak signal-to-noise ratio over initial measurements using the ASOPS technique. This takes place despite the 2056 samples required to obtain the initial results, in comparison to the 256 samples of the present data. Furthermore, the pump and probe beams of the initial results were tuned to the D_2 transition, which has an Einstein coefficient for absorption that is

approximately 12 times larger than that of the $3P_{3/2} \rightarrow 5S_{1/2}$ transition in resonance with the probe beam in the present experiments. The increase in signal-to-noise ratio is attributed to the new trigger arrangement, the detector of Fig. 3, and improved dye laser operation.

Future work with the ASOPS instrument will involve frequency doubling both dye lasers to work in the UV. This will allow us to access the rovibronic transitions of the hydroxyl radical. Atmospheric premixed flames will be used to determine absolute number densities and quenching rates for the hydroxyl radical. The Nd:YAG lasers will then be modified to increase the beat frequency, and ultimately to use third-harmonic mode-locking, so that the data acquisition and averaging system can be optimized to take full advantage of the inherent speed of the ASOPS measurement.

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References

1. P. W. Fairchild, G. P. Smith and D. R. Crosley, *J. Chem. Phys.* **79**, 1975 (1983).
2. J. H. Bechtel and R. E. Teets, *Appl. Opt.* **18**, 4138 (1979).
3. D. R. Crosley, *Opt. Eng.* **20**, 511 (1981).
4. R. P. Lucht, D. W. Sweeney and N. M. Laurendeau, *Combust. Flame* **50**, 189 (1983).
5. J. T. Salmon and N. M. Laurendeau, *Appl. Opt.* **24**, 65 (1985).
6. C. D. Carter, J. T. Salmon, G. B. King and N. M. Laurendeau, *Appl. Opt.* **26**, 4551 (1987).
7. N. S. Bergano, P. A. Jaanimagi, M. M. Salour and J. H. Bechtel, *Opt. Lett.* **8**, 443 (1983).
8. R. Schwarzwald, P. Monkhouse and J. Wolfrum, *Chem. Phys. Lett.* **142**, 15 (1987).
9. Y. Takubo, T. Okamoto, N. Komine and M. Yamamoto, *Jap. J. Appl. Phys.* **26**, 416 (1987).
10. P. A. Elzinga, F. E. Lytle, Y. Jiang, G. B. King and N. M. Laurendeau, *Appl. Spectrosc.* **41**, 2 (1987).

11. P. A. Elzinga, R. J. Kneisler, F. E. Lytle, Y. Jiang, G. B. King and N. M. Laurendeau, *Appl. Opt.* **26**, 4303 (1987).
12. D. Stepowski and M. J. Cottureau, *Appl. Opt.* **18**, 354 (1979).
13. D. Stepowski and M. J. Cottureau, *Combust. Flame* **40**, 65 (1981).
14. R. J. Kneisler, F. E. Lytle, G. J. Fiechtner, Y. Jiang, G. B. King and N. M. Laurendeau, *Optics Letters*, March, 1989.
15. G. J. Fiechtner, Y. Jiang, G. B. King, N. M. Laurendeau, R. J. Kneisler and F. E. Lytle, *Twenty-Second Symposium (International) on Combustion*, The Combustion Institute, Pittsburgh, PA, in press.
16. R. A. Beaman, A. N. Davies, A. J. Langley, W. J. Jones, *Chem. Phys.* **101**, 127 (1986).
17. Y. Takubo, T. Okamoto and M. Yamamoto, *Appl. Opt.* **25**, 740 (1986).